

Lifetime Measurements of the $4f^{14}5d$ Metastable states in Single Ytterbium Ions

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Ytterbium ion has a rich level structure of low-lying states and is one of the widely used ions in experiments with trapped ions. We have measured the radiative lifetimes for the two $4f^{14}5d$ metastable states using single ytterbium ions confined in a rf trap. The obtained lifetimes for $D_{3/2}$ and $D_{5/2}$ states are 52.7(2.4) ms and 7.0(0.4) ms, respectively. The results are in good agreement with the previously measured values but differ significantly from theoretical estimates. The $P_{1/2}$ branching ratio into $D_{3/2}$ has also been obtained from the experiment to be 0.0483 with a 9% uncertainty.

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I. INTRODUCTION

Ytterbium ion is considered the most versatile atomic species used in ion trapping for frequency standards and atomic clocks. This is owing to its rich low-lying energy level structure. Possible frequency standards based on trapped Yb^+ ions range from microwave to infrared to visible frequencies. [1–6] For the same reason, Yb^+ ion is well suited for other possible applications such as studying cavity QED and demonstrating quantum computation using trapped ions.

The energy-level scheme of the low-lying states of Yb^+ ion is shown in Fig. 1. Of particular interest for an optical frequency standard is the $D_{5/2}$ state which decays weakly to the ground state via electric quadrupole transition at 411 nm [4]. The radiative decay lifetime for the $D_{5/2}$ of Yb^+ has been measured before with single trapped ions to be 7.2 ms, which corresponds to a natural linewidth of 22 Hz. [7] Other other hand, the metastable $D_{3/2}$ state, like that in other alkaline-like ions, is often more of an experimental nuisance since the ions are optically pumped into this state by the cooling laser (the $S_{1/2}$ - $P_{1/2}$ transition) and, therefore, a clearing laser is necessary. However, the existence of the $D_{3/2}$ state makes it possible to design the cooling transition to achieve a lower temperature limit. [8] Moreover, the $D_{3/2}$ state in Yb^+ has transitions to several upper mixed states with transition widths of order of MHz, well suited for cavity QED study [9]. The radiative lifetime of the $D_{3/2}$ state of Yb^+ has been measured previously with ion clouds in buffer gases. [10] It was shown that such measurements using ion clouds in buffer gas could lead to considerable error, particularly for long-lived states [11]. Ideally, singly confined ions in UHV should be used for such lifetime measurements to eliminate all possible collisional effects. Unfortunately,

techniques previously used to measure the $D_{3/2}$ lifetimes are not suitable for the Yb^+ $D_{3/2}$ state. A new technique has to be developed.

This paper presents the lifetime measurements on both metastable D states in Yb^+ using trapped single ions. Section II gives a brief description of the experimental setup. Section III and IV describe the measurements for the $D_{3/2}$ and $D_{5/2}$ states respectively. While the measurement for $D_{5/2}$ using quantum jump technique in this work is similar to previous measurements, the $D_{3/2}$ measurement has been carried out with individual ions for the first time in Yb^+ ion using a new measurement technique.

II. EXPERIMENT

A sketch of the experimental setup is shown in Fig. 2. A small Paul-Straubel trap [12] was used to confine single Yb^+ ions. The trap consists of a 1 mm diameter tantalum wire loop with twisted leads as support. The wire diameter is 125 μm . The trap was driven by a 11 MHz rf source. Typical rf voltages applied to the trap were about 700 volts, which produce about 1 MHz axial secular frequency. Because of stray dc fields, the enhanced micromotion of the trapped ions was present. An externally applied compensation was used to reduce the micromotion. However, only one compensation electrode was used roughly along the cooling laser direction since the micromotions perpendicular to the laser beam could not be detected in the present setup.

Ytterbium ions were produced near the trap by directing both ytterbium atoms and the ionizing electron beam towards the trap. The ytterbium source was of natural abundance. Only $^{174}\text{Yb}^+$ was selectively loaded. The selection was through the detuning of the cooling uv laser at 369 nm, which was generated from a frequency-doubled Ti:sapphire laser. Because of the lower-lying $D_{3/2}$ state, a clearing laser is necessary for continuous fluorescence. It can be either a 609 nm laser or a 935 nm laser. For the most of the work presented in this paper, a 609 nm dye laser was used. This transition has a linewidth of about 4 MHz. The micromotion sidebands at 11 MHz can therefore readily resolved. We used the 609 spectrum as a convenient monitor for the micromotion reduction. [13] Both 369 nm and 609 nm laser frequencies were referenced to the absorption lines in Yb^+ discharge lamps. The 369 nm used the direct absorption with a home-made see-through discharge lamp, while the 609 nm absorption was detected using the optogalvanic

spectroscopy technique.

The ion fluorescence photons were collected through an imaging lens and focused into a 50 μm pinhole right in front a solar-blind photomultiplier tube(PMT). A 50% transmitting interference filter of 369 nm was used to cut down the background light. With a single cooled Yb^+ ion, up to 1.6×10^4 photon were counted against less than 100 counts of background. As we will see in the following sections, the large signal to background ratio is crucial for the reliable quantum jump detection at the millisecond scale.

III. MEASUREMENT OF $\text{D}_{3/2}$ LIFETIME

The lifetime of $\text{D}_{3/2}$ in Yb^+ ion has been measured so far only with ion clouds in buffer gas. It is not obvious how the measurement can be done with a single ion because the state is inside the transition cycle for the normal signal detection. A variation of the quantum jump technique was used for measuring the $\text{D}_{3/2}$ state in Ba^+ . [11] For the relative short $\text{D}_{3/2}$ lifetime in Yb^+ , a new approach was called for.

The new measurement method takes advantage of the fact that the branching ratio of the $\text{P}_{1/2}$ decaying to the ground state is large, about 200. In other words, an ion will emit an average of 200 uv photons before getting pumped into the $\text{D}_{3/2}$ state again. If one uses a very short 369 nm pulse, much shorter than the $\text{D}_{3/2}$ state dwell time, then a detection of the 200 fluorescence photons would determine that the ion has decayed to the ground state. A short pulse is also necessary to achieve large enough signal-to-background ratio since the useful signal photons are emitted in the first a few microseconds. In reality, the photon detection efficiency was rather low, about 0.03%. The state detection with a single pulse would be hopeless. Fortunately, one is only interested in the probability in the lifetime measurement and, therefore, can accumulate the photon counts at a fixed wait time over many repetitions. The total accumulated fluorescence count should be proportional to the probability of the ion having decayed within the wait time t_w , i.e. $(1 - e^{-t_w/\tau})$.

The measurement sequence started with a short cooling period with both the cooling and the clearing lasers on. Then the both lasers were turned off with the clearing laser off time trailing the cooling laser. The ion was then left in the ground state. Subsequently, the uv laser was pulsed for a short time to pump the ion into the $\text{D}_{3/2}$ state for the free decay. Photons scattered in this pulse period serve as the $t = 0$ signal. After a given time t_w , another short uv pulse was switched on for the state detection. Finally both lasers were turned on again and the measurement process was repeated. The laser pulse sequence is illustrated in Fig. 3. At the same time, the photon signal of PMT was send to a multi-channel scaler(MCS). The MCS was triggered at a fixed point in

the laser cooling period with a fixed bin width of 10.24 μs . The top trace of Fig. 3 is a sample data collected on MCS. Peak A is the ion fluorescence at the cooling period, peak B is the $t = 0$ signal, and peak D is the $t = t_w$ signal. Pulse C is for the background calibration purpose and will be discussed later. The ratio of the total numbers of the counts under peaks D and B gives the probability for the ion having decayed to the ground state at the time t_w .

It should be pointed out that, for the long-lived lifetime measurement, the laser off-state has to be nearly absolute. Even a very weak leakage of the laser powers is enough to cause the allowed transitions to occur within the time scale under measurement. Therefore, the laser pulses were generated using a custom-made mechanic chopper. Every revolution of the chopper completes one measurement cycle. For convenience, the wait time was varied by changing the chopping frequency f_c . Obviously, all pulse widths change with the chopping frequency. Accordingly, the pulse sequences in Fig.3 are indicated in radians. The corresponding times can be obtained by multiplying the factor of $1/(2\pi f_c)$. The short uv pulses were generated by the holes of about 0.5 mm diameter at 42 mm radius, or 0.012 rad. The separation between the short uv pulses(wait time) is about 6.1 rad. Therefore the timing resolution is 0.2 % regardless of the actual wait time. The fastest chopper speed used was 50 Hz, corresponding to about 20 ms wait time. The wait time t_w for data analysis was directly obtained from the number of bins in the MCS.

There are two background sources needed to be removed from peak signal counts. First is the uniform room light background. It is independent of the chopper state and can be readily subtracted. The second is the stray scattering of the uv laser, which is correlated with the chopping. To remove this scattered photon counts, an additional uv pulse(C in Fig. 3) was used right after the $t = 0$ uv pulse. Since the ion does not decay appreciably in the short time, the collected uv photons are the stray scattering and therefore were subtracted from the peak signal after proper scaling of pulse widths.

In the actual experiment, the wait time was varied from 20 ms to 160 ms. Fig. 4 plots the decay probability vs. the wait time. The error bars indicated are determined from the photon number statistics, i.e. $\delta n = \sqrt{1/n}$. The timing errors are relatively small and not indicated in the figure. A weighted least-squares fitting to the $(1 - e^{-t/\tau})$ function gave 52.7 ± 2.4 ms. Again, the quoted error is derived from the fitting. Typical collisional quenching effect in single ion measurements is on the order of 10^7s^{-1} per Torr [10,11], which is completely negligible in our measurement at 10^{-10} Torr background pressure. The 52.14 ms previously measured in buffer-gas-cooled clouds is consistent with the present single ion result. The theoretical estimate of Fawcett and Wilson [14] is 41 ms while that of Garstang [15] is 74 ms. It should be emphasized here that the single ion measurement here eliminated any need of extrapolations and all known sys-

tematic errors are absent at the quoted uncertainty level.

As a by-product, the $P_{1/2}$ branching ratio to $D_{3/2}$ state can be estimated from our measurement by counting the number of photons emitted during the $t = 0$ pulse before the ion ends in the $D_{3/2}$ state. The overall photon counting efficiency in your system was calibrated by saturating both the cooling and clearing transitions. At the laser intensities 10 times over the saturation intensities for both uv and red lasers, it is easy to show that the $P_{1/2}$ steady-state population is 0.476. The detected resonant and heavily-saturated fluorescence photon count was $16.5k$, i.e. $0.476\eta\gamma_{ps} = 1.65 \times 10^4$. With $\gamma_{ps} = 1.24 \times 10^8$ [16], the detection efficiency was found $\eta = 2.80 \times 10^{-4}$. Given the uncertainties in the degrees of saturation, we assign a 5% uncertainty in the detection efficiency estimate. The averaged photon counts per uv pulse was measured to be 0.0580 ± 0.0038 . Note that this measurement is relatively robust since the average photons scattered per pulse is independent of the laser-induced transition rate and the pulse width. Thus, one obtains the branching ratio to $D_{3/2}$ to be 0.0483 with a 9% uncertainty, agreeing well with the theoretical estimate of 0.048 [17].

IV. MEASUREMENT OF $D_{5/2}$ LIFETIME

The $D_{5/2}$ state is lying below the $P_{1/2}$ states but is not connected to the laser cooling transitions. It serves as the shelving state in the quantum jump experiment. [18] The narrow transition from the ground state is one of the candidates for optical frequency standards. Determining the lifetime, hence the potential oscillator Q , is of great interest.

As a shelving state, the lifetime of the $D_{5/2}$ state can be conveniently determined with the quantum jump technique. In this technique, continuous fluorescence photons are detected when the ion is in the ground state. An excitation of the ion into the $D_{5/2}$ state will quench the fluorescence completely until it decays spontaneously back to the ground state and the fluorescence resumes. The on or off state of the fluorescence signal indicates whether the ion is in the $S_{1/2}$ or $D_{5/2}$ state. The average fluorescence off-time(dark period) gives the lifetime of the $D_{5/2}$ state. The ion excitation into the $D_{5/2}$ state was achieved through the excitation to $P_{3/2}$. For the lack of a 329 nm laser, we used a high current Yb discharge lamp. The lamp could make a shelving rate about 0.3 s^{-1} . The lamp however increased the dark period counts to about 500 when used with a 329 nm interference filter.

There are however two aspects which make the measurement in Yb^+ more difficult than that in other alkaline-like ions. First, below the $D_{5/2}$ state lies an additional $F_{7/2}$ state which is extremely long-lived. In fact, the ion decays into the F state four times more often than back to the ground state. [7] Therefore, an addition clearing laser at 638 nm is necessary. Further more, when the ion signal is off, the ion could either still be in $D_{5/2}$ or

have decayed to the F state. To exclude those events of decaying into the F state, the 638 nm clearing laser was adjusted so that the average dwell time in the F state is much longer than that in the $D_{5/2}$ state. Secondly, the $D_{5/2}$ state lifetime is relatively short. One has to be able to determine the change of the state in millisecond time. We set our detection bin width at 1 ms, which still allowed reliable state detections.

Fig. 5 shows the histogram plot of the ion signal dark period. A least-squares fitting to an exponential decay $e^{-\frac{t}{\tau}}$ yielded $7.0 \pm 0.4 \text{ ms}$, where the error is one standard deviation of the statistic uncertainty from the fitting. The insertion in the figure plots the same histogram at much longer time scale. The “decay” rate of 1.3 s gives the average dwell time of the ion in the F state before repumped out by the 638 nm laser. Subtracting the long time scale decay from the data and re-fitting the short time decay, the resulted change in the $D_{5/2}$ lifetime is insignificant as expected. The measured value agrees well with the previously measured value of 7.2 ms [7] within the quoted experimental errors but significantly differs from a theoretical estimate of 5.7 ms. [14]

V. CONCLUSION

In conclusion, we have measured the natural radiative lifetimes of both $4f^{14}5d$ states in Yb^+ ion. The $D_{3/2}$ experiment has been carried using single ions in UHV for the first time. The obtained values of 52.7 ± 2.4 for $D_{3/2}$ and 7.0 ± 0.4 for $D_{5/2}$ are consistent with previous experimental values but significantly differ from theoretical estimates. The branching ratio of $P_{1/2}$ into $D_{3/2}$ has been found to be 0.0483 with a 9% uncertainty. To our knowledge, this is the first experimental value and agrees well with the theoretical estimate.

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FIG. 1. Yb^+ energy-level scheme of the low-lying states(not to scale).

FIG. 2. Block diagram of the experimental setup.

FIG. 3. The pulse sequence for the $\text{D}_{3/2}$ state lifetime measurement. The time is indicated in radians. The actual time is obtained by multiplying $1/(2\pi f_c)$. The trace of photon counts in the plot was at $f_c = 9.3$ Hz.

FIG. 4. The $\text{D}_{3/2}$ state decay probability vs. the wait time between the uv laser pulses B and D.

FIG. 5. The $\text{D}_{5/2}$ state decay histogram with 1 ms bin. The insertion shows the similar plot for the ion out of the $\text{F}_{7/2}$ state.









